

Claims: #134-173

This set of claims replaces all previous submitted claims

Application number: #09/525,176

Filing Date: 03/14/2000

**Inventors: Savvas Vasileiadis,
Zoe Ziaka**

March 27, 2003

CLAIMS

134. (amended) A process for conducting catalytic reforming of hydrocarbons and alcohols with steam and carbon dioxide for the production of pure hydrogen which includes:

E1
a far outer impermeable hollow tubular cylinder nesting two more concentric permeable tubular cylinders, a next inner and a most inner one, having the most inner permeable cylinder to be nested within the next inner permeable cylinder thus defining three different annular zones including next inner membrane and most inner membrane, with the most inner permeable cylinder to be filled with a reforming catalyst and include gas heating tubes located along the most inner axis, with the catalyst to be in pellet or particle form, with the hydrogen product to be continuously removed via permeation along the most inner membrane wherein the membrane is made from an inorganic or composite material, with the remaining reaction species to partially permeate as well via the most inner membrane, and with the permeated species to be diluted by an inert carrier gas flowing along the next inner annular zone, with hydrogen only to be continuously removed via permeation along the next inner membrane and allow for continuous hydrogen withdrawal out of the most inner catalytic zone and for continuous equilibrium shift of the reactions evolving within this zone, with next inner membrane to be made from a metal or non-porous inorganic material permeable only to hydrogen, with the pure hydrogen to permeate through the next inner membrane and withdrawn along the far outer cylindrical zone.

135. (amended) The process of claim 134 wherein the most inner membrane is made from one or more materials selected from the group consisting of alumina, silica, titania, zirconia, yttria, and the next inner membrane made from one or more materials selected from the group consisting of aluminum carbide and nitride, silicon carbide and nitride, titanium carbide and nitride, zirconium carbide and nitride, tantalum carbide and nitride, palladium, silver, copper, zinc, tantalum, vanadium, tungsten.

136. (amended) The process of claim 134 wherein the feed hydrocarbon or alcohol is a single component or a mixture of components selected from the group consisting of methane, ethane, propane, n-butane, i-butane, methanol, ethanol, propanol, butanol, naphtha, gasoline, natural gas, coal gas containing methane, landfill gas containing methane, flue gas containing methane, biomass and sewage gas containing methane.

137. (amended) The process of claim 134 wherein the combined feed hydrocarbon and carbon dioxide gas mixture is selected from the group consisting of a CH_4 and CO_2 mixture, acidic natural containing CH_4 and CO_2 , coal gas containing CH_4 and CO_2 , landfill gas containing CH_4 and CO_2 , biomass and sewage gas containing CH_4 and CO_2 , flue and waste gas mixture containing CH_4 and CO_2 .

152. (amended) A process for conducting catalytic reforming of hydrocarbons and alcohols with steam and carbon dioxide for production of pure hydrogen which includes:

a far outer impermeable hollow tubular cylinder nesting two more concentric permeable tubular cylinders, a next-inner and a most-inner one, having the most inner

permeable cylinder to be nested within the next inner permeable cylinder thus defining three different annular zones including next inner membrane and most inner membrane, with the annular space between the far outer and next-inner cylinders to be filled with a reforming catalyst, with the catalyst to be in pellet or particle form, with hydrogen to be continuously removed via permeation along the next-inner membrane wherein the membrane is made from an inorganic or composite material, with the remaining reaction species to partially permeate as well via the next inner membrane, and with the permeated species to be diluted by an inert carrier gas flowing along the next inner annular zone, with hydrogen only to be continuously removed via permeation along the most inner membrane in order to allow for continuous hydrogen withdrawal out of the far outer catalytic zone and for continuous equilibrium shift of the reactions evolving within this zone, with the most inner membrane to be made from a metal or non-porous inorganic material, and with the permeate pure hydrogen to withdrawn along the most inner cylindrical zone.

153. (amended) The process of claim 152 wherein the next inner membrane is made from one or more materials selected from the group consisting of alumina, silica, titania, zirconia, yttria, and the most inner membrane made from one or more materials selected from the group consisting of aluminum carbide and nitride, silicon carbide and nitride, titanium carbide and nitride, zirconium carbide and nitride, tantalum carbide and nitride, palladium, silver, copper, zinc, tantalum, vanadium, tungsten.

154. (amended) The process of claim 152 wherein the feed hydrocarbon or alcohol is a single component or a mixture of components selected from the group consisting of methane, ethane, propane, n-butane, i-butane, methanol, ethanol, propanol, butanol, naphtha, gasoline, natural gas, coal gas containing methane, landfill gas containing methane, flue or waste gas containing methane, biomass and sewage gas containing methane.

155. (amended) The process of claim 152 wherein the combined feed hydrocarbon and carbon dioxide gas mixture is selected from the group consisting of a CH_4 and CO_2 mixture, acidic natural gas containing CH_4 and CO_2 , coal gas containing CH_4 and CO_2 , landfill gas containing CH_4 and CO_2 , biomass and sewage gas containing CH_4 and CO_2 , flue and waste gas mixture containing CH_4 and CO_2 .

170. (amended) A process for conducting catalytic hydrocarbon reforming with carbon dioxide, for production of pure hydrogen and carbon dioxide which includes:

a far outer impermeable hollow tubular cylinder nesting two more concentric permeable tubular cylinders, a next-inner and a most-inner one, having the most inner permeable cylinder to be nested within the next inner permeable cylinder thus defining three different annular zones including next inner membrane and most inner membrane, with the annular space between the far outer and next-inner cylinders to be filled with a reforming catalyst in pellet or particle form, with hydrogen and carbon dioxide to be continuously removed via permeation along the next-inner membrane wherein the membrane is made from an inorganic or composite material, with the remaining reaction

species to partially permeate as well via the next inner membrane and with the permeated species to be diluted by an inert carrier gas flowing along the next inner annular zone, with hydrogen and carbon dioxide species to be continuously removed via permeation along the most inner membrane, with the most inner membrane to be made from a polymer or inorganic material which is permeable to both hydrogen and carbon dioxide species, with the permeated binary hydrogen-carbon dioxide mixture to be withdrawn by flowing along the most inner cylindrical zone.

171. (amended) The process of claim 170 wherein the next inner membrane is made from one or more materials selected from the group consisting of alumina, silica, titania, zirconia, yttria, and the most inner membrane made from one or more materials selected from the group consisting of alumina, silica, titania, zirconia, yttria, polyimides, polycarbonates, polybenzimidazoles, polyphosphazenes, polysulfones.

172. (amended) The process of claim 170 wherein the feed hydrocarbon or alcohol is a single component or a mixture of components selected from the group consisting of methane, ethane, propane, n-butane, i-butane, methanol, ethanol, propanol, butanol, naphtha, gasoline, natural gas, coal gas containing methane, landfill gas containing methane, flue and waste gas containing methane, biomass and sewage gas containing methane

173. (amended) The process of claim 170 wherein the combined feed hydrocarbon and carbon dioxide gas mixture is selected from the group consisting of a C_2H_4 and CO_2

E3
mixture, acidic natural gas containing CH_4 and CO_2 , coal gas containing CH_4 and CO_2 ,
landfill gas containing CH_4 and CO_2 , biomass and sewage gas containing CH_4 and CO_2 ,
flue and waste gas mixtures containing CH_4 and CO_2 .
